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^{208}Tl AND ^{24}Na GAMMA SOURCES FOR TRACING SOIL WATER
MOVEMENT WITH DEUTERIUM

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SOIL BRIEF

^{208}Tl and ^{24}Na Gamma Sources for Tracing Soil Water
Movement with Deuterium

^{208}Tl in secular equilibrium with its parent nuclide ^{228}Th is superior to ^{24}Na as a gamma source for tracing downward movement of heavy water (D_2O) in field studies because of its longer half-life.

R. H. Hawkins, R. F. Overman, and J. C. Corey, Savannah River Laboratory, E. I. du Pont de Nemours and Co., Aiken, South Carolina, 29801

ABSTRACT

The D_2O soil water tracer technique is based on the (γ, n) reaction of high-energy gamma photons with deuterium to indicate the tracer's location. The high-energy gamma sources ^{208}Tl and ^{24}Na were compared in their effectiveness to produce neutrons from heavy water (D_2O). In tests with vials of D_2O at various locations in 208-liter (55-gallon) drums of dry and moist soil, ^{24}Na gave 23% higher neutron count rates than ^{208}Tl , but this was more than offset by its shorter effective half-life (15 hours vs. 1.9 years).

Additional Key Words for Indexing: neutrons

^{208}Tl and ^{24}Na Gamma Sources for Tracing Soil Water
Movement with Deuterium¹

R. H. Hawkins, R. F. Overman, and J. C. Corey²

A nondestructive method was described by Haskell and Hawkins (3) for tracing soil water movement in situ using the nonradioactive hydrogen isotope, deuterium, in heavy water (D_2O). Soil water movement can be followed with this method for several weeks because deuterium is stable, and heavy water closely resembles soil water (1,5).

In this method, D_2O is injected at a desired soil depth outside an access tube and its vertical movement is traced by periodically lowering into the access tube a modified soil moisture probe containing a high-energy gamma source (^{24}Na) and a slow-neutron detector. Interaction of gamma photons >2.23 Mev with deuterium nuclei results in the emission of neutrons (2). Some of these neutrons, after being scattered and slowed by light nuclei in the soil, reach the detector in the probe. A neutron count rate peak occurs when the probe is at the depth of maximum D_2O concentration.

A disadvantage of the method, however, is its dependence on ^{24}Na . Although this isotope emits gamma photons of adequate energy (2.75 Mev), its half-life is only 15 hours; thus, the source must be frequently replenished for lengthy investigations. ^{208}Tl was proposed as a long-lived substitute for ^{24}Na (3).

This paper reports a study to determine if ^{208}Tl is a suitable substitute for ^{24}Na . ^{208}Tl emits a 2.61-Mev gamma photon (2) — well

¹Contribution from the Savannah River Laboratory, E. I. du Pont de Nemours and Company, Aiken, South Carolina. The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

²Research Chemists and Research Physicist, respectively.

above the threshold of 2.23 Mev for the (γ, n) reaction — and has an effective half-life of 1.9 years when in equilibrium with its ^{228}Th parent. The ^{228}Th decay chain includes α , β , and γ emitters. Neutron backgrounds of 22 counts per minute per mc of gamma emitter (cpm/mc) in moist soil and 13 cpm/mc in dry soil are attributed to (α, n) reactions with light nuclei such as oxygen.

Because the gamma photons from ^{208}Tl have slightly lower energy than those from ^{24}Na , less energetic neutrons and slightly lower neutron count rates are produced with ^{208}Tl . The two gamma sources were compared experimentally to determine the difference in count rates.

Experimental Materials and Procedure

Deuterium, as 99% D_2O in 100-ml vials, was placed in two 208-liter (55-gallon) drums of Gilead sandy clay. Soil was air-dried, sieved to <2 mm, and packed to a bulk density of 1.5 g/cm^3 with access tubes in place. The soil in one drum was saturated with H_2O , covered with polyethylene sheeting, and allowed to drain for two weeks through small holes in the bottom of the drum. The moisture content of the wetted soil equilibrated at 13% (dry weight basis) and remained there throughout the study. The dry soil (1% moisture) in the second drum was similarly covered with polyethylene sheeting. Tubes protruded through the polyethylene to allow access without exposing the soil to changes in water content.

Each drum contained a center access tube for the probe and 20 similar tubes for D_2O placement, as shown in Figure 1. Four D_2O placement tubes were spaced evenly at each of 5 radial distances from

the center tube. All tubes were 4-cm-I.D. aluminum with 0.22-cm-thick walls, and extended from the bottom of the drum to 5 cm above the soil surface. The two drums were spaced 4 meters apart on a concrete floor.

Heavy water was contained in 100-ml polystyrene vials of 2.56-cm O.D., 12.20-cm length, and 0.13-cm wall thickness. The vials were attached to the lower ends of the aluminum tubes of 3.95-cm O.D. and 0.14-cm wall thickness and 100-cm length. For each drum, four tubes with D₂O vials attached, and 19 similar tubes without vials, were filled with soil corresponding in water content to that in the drum.

For a given determination, one to four of the soil-filled tubes with vials of D₂O were loaded into the access tubes at a single radial distance in one drum. Soil-filled tubes without vials were loaded into the remaining tubes to minimize void space. The probe containing the detector and source was placed in the center access tube to measure the neutron count rate.

The vertical positions of the probe and D₂O vials were varied to obtain the maximum count rates. This probe position, 10 cm above the bottom of the center access tube with the D₂O vials at the bottom of other access tubes, was used throughout the study.

Probe and Scaler

A modified Nuclear-Chicago P-19 depth moisture probe was used. The original BF₃ neutron detector tube in the probe was replaced with a ³He-filled tube³ to increase neutron detection efficiency. The gamma source was in the lower part of the probe housing.

³Type 252, LND Inc., Oceanside, N. Y.

The detector tube was gamma-shielded by a lead cylinder 7-cm long and 3.68-cm in diameter. Neutron counts were registered on a Nuclear-Chicago 3600A portable scaler operated on 60-cycle, 120-volt alternating current. Two-minute counts, corrected for background, were used throughout the study.

Gamma Sources

The ^{24}Na source consisted of 2.3 g of dry Na_2SiO_3 encapsulated in glass and sealed in an aluminum cylinder of 1.59-cm diameter and 2.80-cm length. The cylinder and its contents received 10^{10} neutrons/cm² in a nuclear reactor. At the completion of this irradiation the source contained 20.5 mc of ^{24}Na , based on measurement of its high-energy gamma radiation.

The ^{208}Tl source (4) was made by electrodepositing the parent nuclide, ^{228}Th , into a bismuth matrix. Bismuth was used to reduce neutron production from (α, n) reaction with light nuclei such as oxygen. The source was encapsulated in a stainless steel cylinder of 3.0-cm diameter and 3.5-cm length. After ^{208}Tl had equilibrated with ^{228}Th (about 5 weeks), measurement of the high-energy gamma radiation showed the source contained 2.70 mc of ^{208}Tl .

RESULTS AND DISCUSSION

Neutron count rates measured with D_2O at distances of 7.0, 9.5, and 14.5 cm are given in Figure 2. Count rates at 19.5 and 22.0 cm are not reported because they were too low to be reliable. The ^{24}Na data were corrected for radioactive decay at 15-minute intervals.

Decay correction of ^{208}Tl data was considered unnecessary due to the 1.9-year half-life of this source.

The data in Table 1 and Figure 2 show that higher neutron count rates were obtained per mc of ^{24}Na than of ^{208}Tl when other factors were constant. This was expected because the ^{24}Na gamma photon has 0.14-Mev greater energy than that of ^{208}Tl . However, the small differences between count rates for ^{24}Na and ^{208}Tl were not of major concern in selecting a source for D_2O soil water tracing.

The highest neutron count rates were obtained with ^{24}Na , in moist soil, and at the closest (7-cm) distance between the probe and the largest amount of D_2O (Table 1 and Figure 2). This result was expected because these conditions are most favorable to neutron production and detection. Slopes of the lines in Figure 2 are proportional to the neutron count rates per mc of gamma emitter. Comparison of these slopes reveals greater dependence on soil water content, and distance between D_2O and probe, than on the type of gamma source ^{208}Tl or ^{24}Na .

The ratios $\frac{\text{cpm/mc with } ^{208}\text{Tl}}{\text{cpm/mc with } ^{24}\text{Na}}$ were computed (Table 2) to provide a better comparison between the relative neutron productivity from D_2O by the two gamma sources. No systematic variation appears in these ratios as a result of distance between probe and D_2O or of soil water content. This result was expected because neutron count rates with both ^{208}Tl and ^{24}Na should be affected equally by these factors. In contrast, the general increase in this ratio with increasing volume of D_2O was not expected; the authors have no current explanation for this observation.

Based on the mean overall count rate ratio of 0.79 (the most useful finding of this study), 1.27 mc of ^{208}Tl will produce the same count rate of neutrons from D_2O as 1.00 mc of ^{24}Na .

SUMMARY AND CONCLUSIONS

1. A ^{208}Tl - ^{228}Th gamma source is satisfactory for the (γ, n) reaction with D_2O soil moisture tracer.
2. A ^{208}Tl - ^{228}Th gamma source produced neutron count rates from D_2O averaging 79% of those produced with an equal activity of ^{24}Na .
3. The 1.9-year half-life of a ^{208}Tl - ^{228}Th gamma source offers a significant advantage over the 15-hour half-life of a ^{24}Na gamma source, and more than compensates for the slightly lower neutron yield from D_2O with ^{208}Tl .

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Figure 1. Access Tube Arrangement in Soil Drums.

Figure 2. Neutron Count Rates from (γ, n) Reaction With Deuterium Using ^{24}Na and ^{208}Tl Gamma Sources.

Table 1. Count rate of neutrons (corrected for background) for different volumes of D₂O, in dry and moist soil, at different distances from the source-detector probe containing ²⁰⁸Tl or ²⁴Na.

Gamma Source in Probe	Water Content of Soil, g/g	D ₂ O Volume, ml	Count Rate, cpm/mc		
			7 cm*	9.5 cm*	14.5 cm*
²⁴ Na	0.13	100	190	70	15
		200	324	143	29
		300	443	200	39
		400	590	251	53
²⁴ Na	0.01	100	104	48	13
		200	189	80	25
		300	242	114	34
		400	309	148	46
²⁰⁸ Tl	0.13	100	130	49	10
		200	248	96	21
		300	363	164	33
		400	530	215	52
²⁰⁸ Tl	0.01	100	94	31	10
		200	132	60	22
		300	210	103	29
		400	270	134	37

* Distance between D₂O and probe.

Table 2. Ratios of $\frac{\text{cpm/mc with } ^{208}\text{Tl}}{\text{cpm/mc with } ^{24}\text{Na}}$ from different volumes of D_2O at different distances from the source-detector probe in moist or dry soil.

Water Content of Soil, g/g	D_2O Volume, ml	Count Rate, cpm/mc		
		7.0 cm*	9.5 cm*	14.5 cm*
0.13	100	0.68	0.71	0.67
	200	0.77	0.67	0.71
	300	0.82	0.82	0.85
	400	0.90	0.86	0.98
0.01	100	0.90	0.64	0.76
	200	0.70	0.75	0.65
	300	0.87	0.91	0.84
	400	0.87	0.90	0.80

Mean overall ratio: 0.79

* Distance between D_2O and probe.

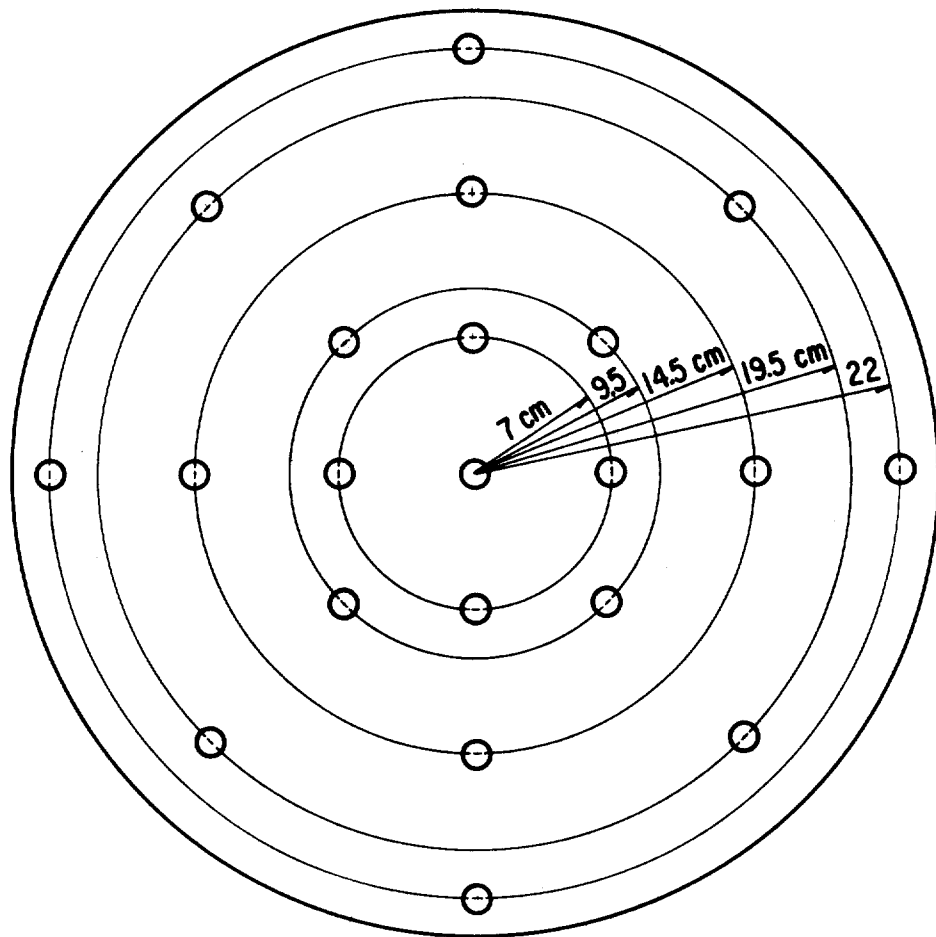


FIG. 1 ACCESS TUBE ARRANGEMENT IN SOIL DRUMS

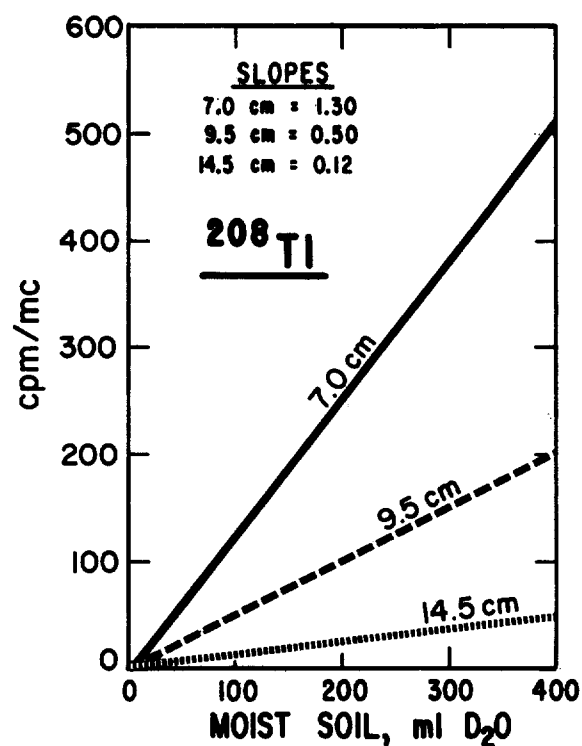
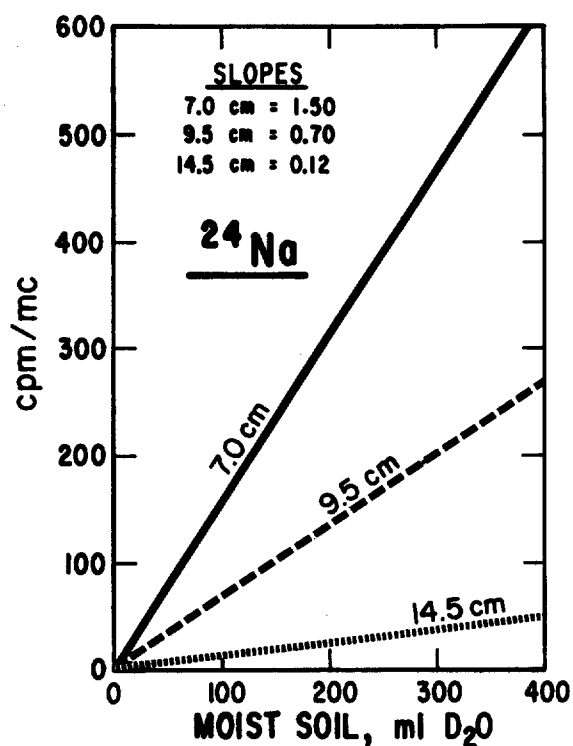
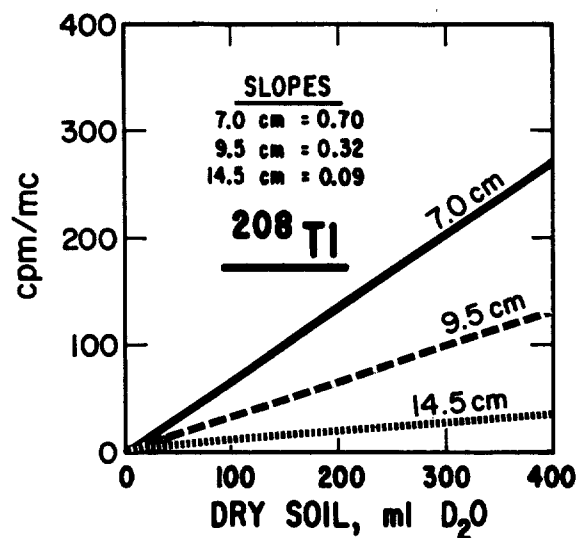
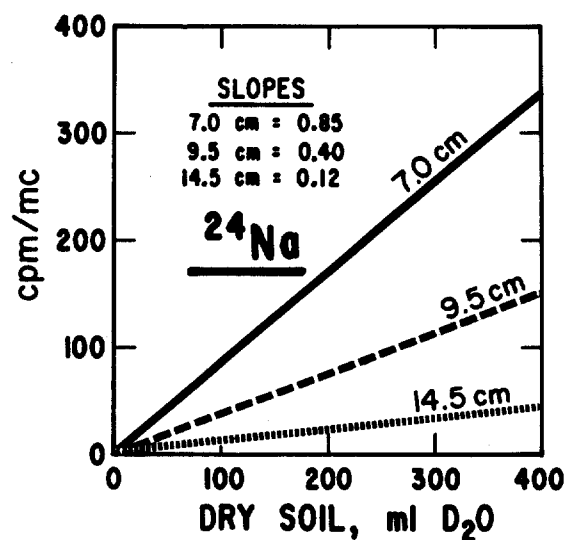


FIG. 2 NEUTRON COUNT RATES FROM (γ, n) REACTION WITH DEUTERIUM USING ²⁴Na AND ²⁰⁸Tl GAMMA SOURCES

^{208}Tl AND ^{24}Na GAMMA SOURCES FOR
TRACING SOIL WATER MOVEMENT WITH DEUTERIUM

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SOIL BRIEF

^{208}Tl and ^{24}Na Gamma Sources for Tracing Soil Water Movement with Deuterium

^{208}Tl in secular equilibrium with its parent nuclide ^{228}Th is superior to ^{24}Na as a gamma source for tracing downward movement of heavy water (D_2O) in field studies because of its longer half-life.

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ABSTRACT

In situ tracing of soil water movement with the D_2O - ^{24}Na method has been limited by the need to reactivate the ^{24}Na gamma source frequently (half-life of 15 hours) in a nuclear reactor. Our laboratory study showed ^{208}Tl produced the same neutron count rates as did ^{24}Na from vials of D_2O in dry and moist soil. The 1.9 years effective half-life of ^{208}Tl in equilibrium with its parent nuclide ^{228}Th is a significant advantage of ^{208}Tl over ^{24}Na . Recent commercial availability of ^{208}Tl sources now make *in situ* D_2O tracing of soil water feasible for investigators not having access to a nuclear reactor.

Additional Key Words for Indexing: neutrons

^{208}Tl AND ^{24}Na GAMMA SOURCES FOR
TRACING SOIL WATER MOVEMENT WITH DEUTERIUM¹

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A nondestructive method for tracing the vertical movement of soil water *in situ* using the nonradioactive hydrogen isotope (deuterium) as the oxide, heavy water (D_2O), was reported by Haskell and Hawkins (3). The method consisted of injecting D_2O (usually 400 ml) at a desired depth in soil close to an access tube. The vertical movement of the D_2O -tagged soil water was then traced by determining its position as often as desired by scanning the access tube with a probe and scaler. The probe contained a high energy gamma source (^{24}Na) and a slow-neutron detector. The method was based on the interaction, with deuterium nuclei in the D_2O , of gamma photons >2.23 Mev from the source to produce photo-neutrons (2), some of which collide with light nuclei in the soil and are back-scattered to the detector as slow neutrons. The highest count rate of slow neutrons occurs at the depth of the D_2O -tagged water, and periodic scans trace the vertical movement of this count rate peak.

Deuterium is a good soil water tracer (1,8) and the tracing method provides opportunities for field studies of soil water movement which cannot be made by other methods. Use of the method, however, has been limited by the short half-life (15 hours) of ^{24}Na . Lengthy studies require frequent reactivation of a ^{24}Na source necessitating a nuclear reactor, which is not available to many investigators.

¹Contribution from the Savannah River Laboratory, E. I. du Pont de Nemours and Company, Aiken, South Carolina.

The information contained in this article was developed during the course of work under Contract AT(07-3)-1 with the U. S. Atomic Energy Commission.

²Research Chemists and Research Physicist, respectively.

^{208}Tl has been proposed as a long-lived substitute for ^{24}Na (3) but satisfactory ^{208}Tl sources have only recently become available. This paper describes a study comparing ^{208}Tl and ^{24}Na gamma sources for soil water tracing with deuterium.

EXPERIMENTAL MATERIALS AND PROCEDURE

Gamma Sources

^{208}Tl emits a gamma photon of 2.61 Mev energy; that of ^{24}Na is 2.75 Mev (2). Both energies are well above the threshold of 2.23 Mev for the (γ, n) reaction with deuterium. A ^{208}Tl source is actually ^{228}Th in which ^{208}Tl , a radioactive daughter, has reached secular equilibrium. Although ^{208}Tl has a half-life of only 3 minutes, the "effective" half-life of a ^{228}Th - ^{208}Tl source is that of ^{228}Th , 1.9 years.

The ^{208}Tl source was made by electrodepositing the parent nuclide, ^{228}Th , into a bismuth matrix (7). Bismuth was used to reduce the light elements (especially oxygen) in the source, because light nuclei cause an unwanted neutron background through an (α, n) reaction. The source was sealed into a stainless steel cylinder of 3.0-cm outside diameter and 3.5 cm in length. After ^{208}Tl reached secular equilibrium with ^{228}Th , requiring 3 weeks (6), measurement of the high energy gamma radiation showed there were 2.7 mc of ^{208}Tl .

The ^{24}Na source consisted of 2.3 g of dry Na_2SiO_3 encapsulated in glass and sealed in an aluminum cylinder of 1.6-cm outside diameter and 2.8 cm in length. The sodium in the source was activated to ^{24}Na with a total neutron flux of 10^{10} neutrons/cm² in a nuclear reactor. Measurement of the high energy gamma radiation from the source showed 20.5 mc of ^{24}Na shortly after activation.

Decay correction was computed from this time.

Probe and Scaler

A modified Nuclear-Chicago P-19 depth moisture probe contained both the slow neutron detector and the gamma source (^{208}Tl or ^{24}Na). The BF_3 slow neutron detector tube originally in the probe was replaced with a ^3He -filled slow neutron detector tube³ to increase detection efficiency.

The gamma source was located below the detector tube and 2 cm above the bottom of the 47.8-cm-long probe housing. A cylindrical lead shield 3.68 cm in diameter and 7 cm long separated the gamma source from the detector tube to reduce gamma-induced background. Neutron counts were recorded on a Nuclear-Chicago 2800A portable scaler. Two-minute counts were made throughout the study.

Soil

Gilead sandy clay was air dried, sieved to <2 mm, and packed to a bulk density of 1.5 g/cm^3 in two 208-liter (55 gallon) steel drums 56 cm in diameter and 83 cm long, with access tubes in place. The soil in one drum was saturated with H_2O , covered with polyethylene sheeting, and allowed to drain for two weeks through perforations in the bottom of the drum. The water content of the wetted soil remained at 0.13 g/g (dry weight basis) throughout the study. The air dry soil in the second drum was similarly covered with polyethylene sheeting and remained at 0.01 g/g throughout the study.

³Type 252, LND Inc., Oceanside, N. Y.

Geometry

Each drum contained 21 access tubes consisting of a center tube into which the source-detector probe was placed, and 20 similar tubes for placement of D₂O vials. The access tube arrangement is shown in Figure 1. It can be seen from the top view that at each of the five radial distances four access tubes for D₂O placement were uniformly spaced around the center tube. All access tubes were aluminum with 4-cm inside diameters, 0.22-cm wall thicknesses, and extended from the bottom of the drum to 5 cm above the soil surface. The two drums were placed upright, 4 meters apart on a concrete floor.

Deuterium for the (γ ,n) reaction with the ²⁰⁸Tl radiation was in the form of 99% D₂O in 100-ml polystyrene vials. The vials were 2.56 cm in outside diameter, 12.20 cm long, and had wall thicknesses of 0.13 cm. Each vial was sealed and attached to the bottom of an aluminum tube of 3.95 cm outside diameter, 100 cm length and 0.14-cm wall thicknesses. Eight such tubes with D₂O vials attached, and 40 tubes without D₂O vials, were packed with soil as used in the drums (24 moist soil, 24 dry soil). These were placed in the access tubes of the drums as needed.

Count Rate Measurements

For all measurements, the probe was positioned in the center access tube of a drum of soil, 10 cm above the bottom of the drum. Lower positioning was prevented by a damaged center access tube in one drum. D₂O vials were at the bottom of their access tubes, resting on the bottom of the drum. Soil-filled tubes occupied the access tube space above the D₂O vials and also filled all other access tubes during count rate measurements. Actual distances from the center of the gamma source to centers of D₂O vials in the various radial

positions are shown in Figure 1.

Prior to each count rate measurement with D₂O, duplicate two-minute background counts were made with soil-filled tubes occupying all access tubes and with no D₂O vials present. From one to four soil-filled tubes at a single radial distance were next replaced with soil-filled tubes attached to 100 ml vials of D₂O. Duplicate two-minute counts were then made before repeating background counting and proceeding with a different D₂O configuration. All count rates, including background, are expressed as counts per minute per millicurie (cpm/mc) of the gamma source used to facilitate direct comparisons between results obtained with different mc amounts of ²⁰⁸Tl and ²⁴Na.

RESULTS AND DISCUSSION

Neutron count rates obtained when the probe and D₂O vials were separated by distances of 10.4, 12.2, and 16.4 cm are given in Figure 2. Count rates obtained when the probe and D₂O vials were separated by distances of 21.0 and 23.3 cm were low and unreliable. Satisfactory results at the greater distances could have been obtained with longer counting times, larger gamma sources, or larger amounts of D₂O, but were not essential for comparing the two sources. Count rates obtained with ²⁴Na were corrected for radioactive decay of this source at 15-minute intervals. No correction was made for decay of the ²⁰⁸Tl source because its decay during the three days of use was less than 0.5%.

The results in Figure 2 show that under comparable conditions of soil moisture, distance, and D₂O amount, similar neutron count rates were measured per mc of ²⁰⁸Tl and of ²⁴Na. This is the most significant result of this study and provides a basis for the comparative evaluation of the two gamma sources.

The similar neutron count rates obtained with the two gamma sources under a range of conditions indicates that the difference of 0.14 Mev between their high energy gamma photons has a negligible effect on neutron production from deuterium in soil studies.

Highest neutron count rates with either ^{208}Tl or ^{24}Na were obtained with four 100-ml vials of D_2O at 10.4 cm from the probe in the drum of moist soil. This was anticipated because these conditions placed our maximum amount of D_2O at our minimum distance from the probe in soil which was most favorable for neutron scattering. Conversely, lowest count rates shown above background with either ^{208}Tl or ^{24}Na were obtained with one 100-ml vial (our minimum amount) of D_2O at 16.4 cm distance (our maximum shown) from the probe in the drum of dry soil which was least favorable for neutron scattering.

Background counts result from several reactions and interactions including (α, n) reactions with light nuclei within the sealed ^{208}Tl source, (γ, n) reactions with natural deuterium in the soil and soil water, gamma effect on the detector (sometimes called "gamma noise"), and cosmic radiation. Background count rates cannot be properly expressed as cpm/mc of gamma emitter because some of the contributions to background are not proportional to the size of the gamma source. Instead representative background count rates and the conditions under which they were measured will be given.

Representative background count rates measured in the drum of moist soil were 166 cpm with 8.8 mc of ^{24}Na and 58 cpm with 2.8 mc of ^{208}Tl . Comparable background count rates measured in the drum of dry soil were 61 cpm with 7.7 mc of ^{24}Na and 33 cpm with 2.7 mc of ^{208}Tl . Recognizing that a part of the background count rate results from the amount of gamma emitter present, it is noted

that backgrounds measured with ^{208}Tl were slightly lower per mc of gamma emitter than were those measured with ^{24}Na . This suggests that the backgrounds associated with our ^{208}Tl source had no large contribution from (α, n) reactions with associated light nuclei within the source.

Our 2.7 mc ^{208}Tl source was adequate for the comparative measurements made under the controlled conditions of this study. The source, however, was considered too small for satisfactory field use. Approximately 50 mc of ^{208}Tl are considered adequate for field use and should permit accurate soil water tracing with an injection of 400 ml of D_2O . (A 50 mc ^{24}Na source would require replenishment or reactivation daily to maintain >25 mc, but a 50 mc ^{208}Tl source could be used for nearly two years before decaying to 25 mc).

In situ tracing of soil water movement differs from the measurements in drums of soil reported here primarily in that the D_2O is injected into the soil and is not confined in vials. Injected tracer becomes increasingly diffuse with time and with movement of soil water, resulting in a decreasing count rate. To compensate for the greater counting error as count rates decline, longer counting times may be necessary (5). Excessive length of counting time and flattening of the tracer peak eventually dictate when a new volume of D_2O is required at another depth or access tube site to continue tracing.

The recently announced availability of ^{208}Tl sources from a commercial producer⁴ and the continuing sale of D_2O for research use⁵ offer increased opportunity for investigating soil water movement. In addition to the demonstrated effectiveness of the D_2O tracer method for measuring downward movement of soil water at shallow depths (3,4), there are numerous other possible applications. These applications include measurement of the upward component of

soil water movement at shallow depths, measurement of both upward and downward movement of soil water immediately above the water table, measurement of soil water flow parallel to the surface of steep slopes, and evaluation of design and construction of water holding structures by measuring water movement in the underlying soil.

⁴Amersham Searle Corp., Des Plaines, Illinois.

⁵U. S. Atomic Energy Commission, Savannah River Plant, Aiken, S.C.

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LIST OF FIGURES

Figure 1. Access tubes in drums of soil.

Figure 2. Neutron Count rates with four levels of D₂O at three distances from ²⁰⁸Tl or ²⁴Na in dry or moist soil.

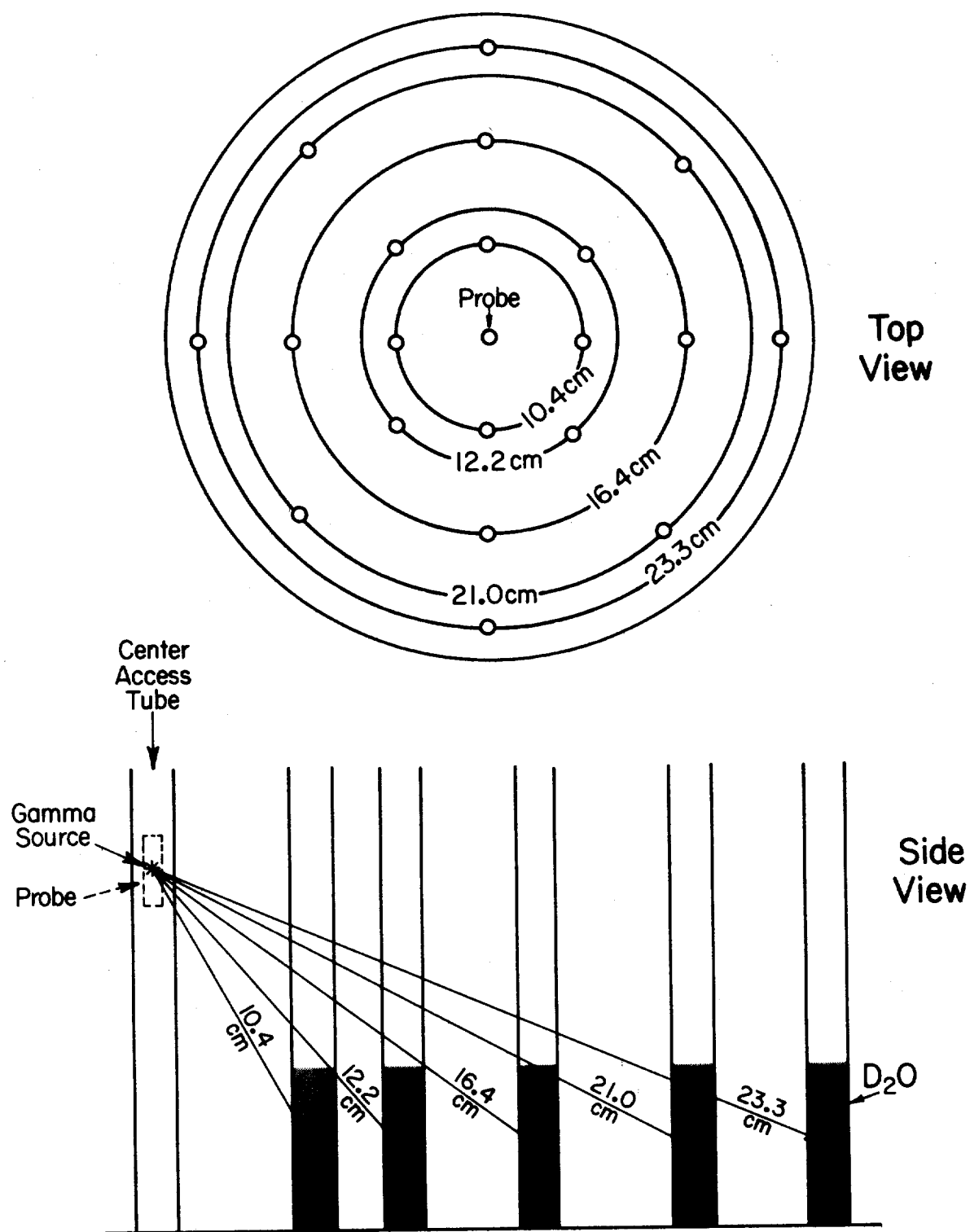


FIG. 1 ACCESS TUBES IN DRUMS OF SOIL

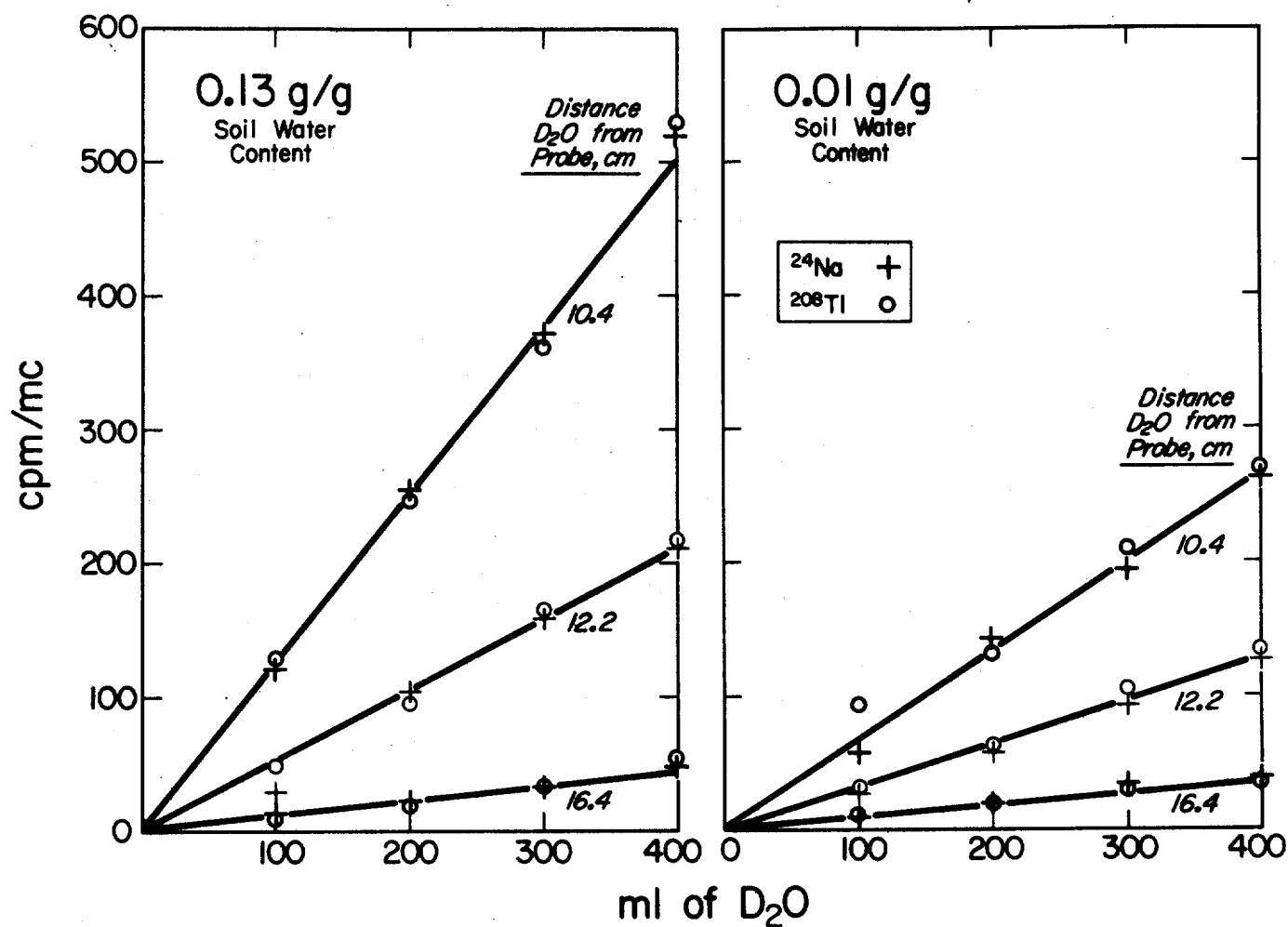


FIG. 2 NEUTRON COUNT RATES WITH FOUR LEVELS OF D₂O AT THREE DISTANCES FROM ²⁰⁸Tl OR ²⁴Na IN DRY OR MOIST SOIL